

# Nuclear spin ferromagnetic phase transition in an interacting 2D electron gas

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Electrons in a two-dimensional semiconducting heterostructure interact with nuclear spins via the hyperfine interaction. Using a Kondo lattice formulation of the electron-nuclear spin interaction, we show that the nuclear spin system within an interacting two-dimensional electron gas undergoes a ferromagnetic phase transition at finite temperatures. We find that electron-electron interactions and non-Fermi liquid behavior substantially enhance the nuclear spin Curie temperature into the  $mK$  range with decreasing electron density.

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The use of the electron spin as a qubit for quantum computing relies on the ability to coherently control single electron spins in semiconductor quantum dots [1]. Over the last years much progress has been made for dots in GaAs semiconductors, where single spin lifetimes have been measured to range far into the ms-range [2, 3, 4], and where coherent manipulation of single- and two-spin states was successfully implemented [5, 6]. Still, a major obstacle to further progress is the comparatively short spin decoherence time in these materials, ranging from 100 ns in bulk [7] to  $\mu s$  in dots [5]. The main source of decoherence for a single electron spin confined to a GaAs dot is coming from the contact hyperfine interaction with the surrounding nuclear spins [8, 9, 10]. Several ways to overcome this problem have been proposed such as spin echo techniques [5, 9], projection of the nuclear spin state [9] or polarization of the nuclear spins [8, 9, 10, 11]. However, in order to extend the spin decay time by one order of magnitude, a polarization of above 99% is required [9], which is still far away from the 60% so far reached in quantum dots via optical pumping [12]. One way to overcome this problem would be that nuclear spins become fully polarized at low enough temperatures, without any external magnetic field or optical pumping. This is the case if the nuclear spins undergo a ferromagnetic phase transition at a finite Curie temperature  $T_c$ . Quite remarkably, the possibility of such a nuclear-spin phase transition to occur in a metal was studied more than sixty years ago by Fröhlich and Nabarro (FN) [13]. Using a Weiss mean field argument they showed that the Curie temperature  $T_c$  of nuclear spins in a three dimensional (3D) metal becomes

$$k_B T_c \sim \frac{A^2}{8E_F}, \quad (1)$$

where  $A$  denotes the hyperfine coupling strength between the nuclear and electron spin and  $E_F$  the Fermi energy. For a typical metal,  $T_c$  is of the order of micro-Kelvin or less. However, for a two-dimensional electron gas (2DEG) in GaAs semiconductors, Eq. (1) would predict nuclear ferromagnetism with  $T_c \sim 1mK$ , which is

surprisingly high

However, the direct use of Eq. (1), which was derived for a bulk metal, to a 2DEG in a semiconductor is very problematic. The purpose of this letter, therefore, is to reconsider this issue for a 2DEG and to estimate the nuclear spin Curie temperature. Our analysis below will be based on the Kondo lattice model [14], where we integrate out the electron degrees of freedom to derive an effective spin Hamiltonian whose exchange is given in terms of the static electronic spin susceptibility  $\chi_s(q)$ . Using a spin-wave analysis, we will show that the electron-electron (e-e) interactions in the 2DEG and the induced non-Fermi liquid behavior in  $\chi_s(q)$  [15, 16, 17, 18] ultimately enables a ferromagnetic phase transition of the nuclear spins. For sufficiently strong interactions and/or low electronic densities (with the dimensionless interaction parameter  $r_s \sim 5 - 10$ ) the Curie temperature can be pushed into the milli-Kelvin regime, and thus, the phase transition should become accessible experimentally.

**Model Hamiltonian.** In order to study an interacting 2DEG coupled to nuclear spins within the 2DEG, we adopt a tight-binding representation in which each lattice site contains a single nuclear spin and electrons can hop between neighboring sites. The Hamiltonian describing such a system reads

$$H = H_0 + \frac{1}{2} \sum_{j=1}^N A_j c_{j\alpha}^\dagger \vec{\sigma}_{\alpha\beta} c_{j\beta} \cdot \vec{I}_j = H_0 + H_n, \quad (2)$$

where  $H_0$  denotes the conduction electron Hamiltonian and  $H_n$  the electron-nuclear spin hyperfine interaction.  $H_0$  can be rather general and includes e-e interactions. In Eq. (2),  $c_{j\alpha}^\dagger$  creates an electron at the lattice site  $\vec{r}_j$  with spin  $\alpha$  and  $\vec{\sigma}$  represent the Pauli matrices. We have also introduced  $\vec{I}_j$  the nuclear spin located at the lattice site  $\vec{r}_j$ , and  $A_j$  the hyperfine coupling constants between the electron and nuclear spins at site  $\vec{r}_j$ . The electron spin operator is defined by  $\vec{S}_j = \frac{1}{2} c_{j\alpha}^\dagger \vec{\sigma}_{\alpha\beta} c_{j\beta}$ .  $N$  denotes the total number of sites on the 2D lattice. In our formulation, the nuclear spin density is  $n_s = a^{-2}$  where  $a$  is the lattice spacing. From here on, we assume

$A_j = A > 0$  which means we assume the hyperfine interaction to be the same for all atoms that constitute the heterostructures (typically Ga and As). We also neglect direct dipolar interactions between the nuclear spins which is in general smaller than the indirect interaction as we will see. This amounts to assume that the dipolar interaction energy scale  $E_{dip}$  is among the smallest one and particularly that  $k_B T \gg E_{dip}$ , where  $T$  is the temperature. This assumption is crucial since it allows us to focus on the nuclear spins which are within the 2D electron gas thickness (in growth direction) and justifies our 2D description [19].

The general Hamiltonian in Eq. (2) is the well-known 2D Kondo lattice Hamiltonian (KLH), though  $H_0$  contains also e-e interactions. The regime we are interested in corresponds to the weak Kondo coupling regime in the sense that  $A \ll E_F$ , where  $E_F$  is the Fermi energy. The KLH has been introduced to describe various physical properties of heavy-fermion materials [14, 20], and more recently also of ferromagnetic semiconductors [21].

Before turning to the extended system let us briefly consider the special case of a single electron confined to a quantum dot which interacts typically with  $10^6$  nuclear spins [9, 10]. This case can be described by the above KLH by allowing in  $H_0$  for a confinement potential for the dot, which provides the largest energy scale. Indeed, we can then project  $H_n$  into the ground state of  $H_0$ , and the hyperfine Hamiltonian then takes the known central spin form  $H = \sum_i \tilde{A}_i \vec{S}_e \cdot \vec{I}_i$  [9, 10], where  $\vec{S}_e$  is the single electron spin, and  $\tilde{A}_i = A |\psi(\vec{r}_i)|^2$  the non-uniform coupling constant with  $\psi(\vec{r}_i)$  the electronic ground state wave function at site  $\vec{r}_i$ . The reformulation of the central spin problem in terms of the KLH should be particularly useful for numerical evaluations.

To continue with the general case, it is convenient to go to Fourier space and rewrite  $H_n$  in Eq. (2) as  $H_n = \frac{A}{N} \sum_{\vec{q}} \vec{S}_{\vec{q}} \cdot \vec{I}_{\vec{q}}$ , where  $\vec{I}_{\vec{q}} = \sum_j e^{-i\vec{q} \cdot \vec{r}_j} \vec{I}_j$  is the Fourier transform of  $\vec{I}_j$ , and similarly for  $\vec{S}_{\vec{q}}$ . Since  $A$  is a small energy scale in our case, we may perform a Schrieffer-Wolff (SW) transformation in order to eliminate terms linear in  $A$ , and thereby integrate out the electronic degrees of freedom. Keeping the lowest order terms in  $A^2$  of the SW transformation, we are left with an effective Hamiltonian  $H_{eff} = H_0 - \frac{1}{2} [S, [S, H_0]]$ .  $S$  is defined by  $H_n + [S, H_0] = 0$ , which is solved as  $S = L_0^{-1} H_n$  where  $L_0$  is the Liouvillian. Let us define  $U = \frac{1}{2} [S, [S, H_0]]$  which can be rewritten as  $U = \frac{1}{2} [L_0^{-1} H_n, H_n]$ . Using an integral representation for  $L_0$ , one obtains  $U = -\frac{i}{2} \int_0^\infty dt e^{-\eta t} [H_n(t), H_n]$ , where  $\eta \rightarrow 0^+$  ensures convergence. We next take the equilibrium expectation value over electronic degrees of freedom, denoted by  $\langle \dots \rangle$ . The only assumptions we make are  $\langle S_i^x \rangle = \langle S_i^y \rangle = 0$ , and translational invariance in the 2DEG. We then get

$$\langle U \rangle = \frac{A^2}{8n_s} \sum_{\vec{q}} I_{\vec{q}}^\alpha \chi_{\alpha\beta}(q) I_{-\vec{q}}^\beta, \quad (3)$$

where  $\chi_{\alpha\beta}(q) = -i \int_0^\infty dt e^{-\eta t} \langle [S_{\vec{q}}^\alpha, S_{-\vec{q}}^\beta] \rangle$ , and where summation over the spin components  $\alpha, \beta = x, y, z$  is implied. If we also assume  $\langle S_i^z \rangle = 0$ , then  $\chi_{\alpha\beta}(q) = \delta_{\alpha\beta} \chi_s(q)$ , where  $\chi_s(q)$  is the electronic spin susceptibility in the static limit. We stress that Eq. (3) is rather general and requires only weak assumptions on  $H_0$ . In real space we have  $\langle U \rangle = -\frac{1}{2} \sum_{\vec{r}, \vec{r}'} J_{\vec{r}-\vec{r}'}^{\alpha\beta} I_{\vec{r}}^\alpha I_{\vec{r}'}^\beta$ , where  $J_{\vec{r}}^{\alpha\beta} = -(A^2/4n_s) \chi_{\alpha\beta}(\vec{r})$  is the effective exchange coupling. The nuclear spins  $\vec{I}_{\vec{r}}$  are therefore interacting with each other, this interaction being mediated by the conduction electrons. This is nothing but the standard Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, which, as we shall see, can be substantially modified by e-e interactions compared to the free electron case.

Let us first analyze the case of non-interacting electrons. In this case,  $\chi_s$  coincides with the usual density-density response (Lindhard) function  $\chi_0$  [22]. We first perform a mean field analysis. The Weiss mean field theory predicts a Curie temperature

$$T_c = -\frac{I(I+1)}{3k_B} \frac{A^2}{4n_s} \chi_0(q=0), \quad (4)$$

where  $I$  is the nuclear spin value. In 2D,  $\chi_0(q=0) = -N_e = -m^*/\pi$ , where  $N_e = n_e/E_F$  is the electronic density of states, and  $m^*$  is the effective electron mass in a 2DEG (we set  $\hbar = 1$ ). For a 3D bulk metal with one conduction electron per nucleus, the ratio  $n_e/n_s \sim 1$  and we recover the result in Eq. (1) derived more than sixty years ago by Fröhlich and Nabarro [13]. For a 2D metal, the Weiss mean field theory predicts  $k_B T_c = I(I+1)A^2/12E_F$ . For a 2D semiconductor, however, the ratio  $n_e/n_s$  is much smaller than 1. With typical values for GaAs heterostructures,  $I = 3/2$ ,  $A \sim 90 \mu\text{eV}$  and  $a \sim 2\text{\AA}$  [9], we estimate  $T_c \sim 1 \mu\text{K}$ , which is very low. (For such low  $T_c$ 's, ignoring nuclear dipole-dipole interactions from the start would not be valid.) However, this estimate is just based on the simplest mean field theory and, moreover, does not include the effect of e-e interactions.

We shall now go beyond above mean field approximation. For this we assume that the ordering (if it takes place) leads to a ferromagnetic phase where the collective low-energy excitations are given by spin waves. Then, we define the Curie temperature  $T_c$  as the temperature at which the magnetic order is destroyed by those spin waves. This procedure is equivalent to the Tyablikov decoupling scheme [24]. The dispersion relation of the spin wave (or magnon) reads

$$\omega_q = I(J_0 - J_q) = I \frac{A^2}{4} a^2 (\chi_s(q) - \chi_s(0)), \quad (5)$$

where  $J_q$  is the Fourier transform of  $J_{\vec{r}}$ . The magnetization  $m$  per site at finite  $T$  is  $m(T) = I - \frac{1}{N} \sum_{\vec{q}} n_q$ , where  $n_q = (e^{\omega_q/k_B T_c} - 1)^{-1}$  is the magnon occupation number. The Curie temperature  $T_c$  follows then from the vanishing of the magnetization, i.e.  $m(T_c) = 0$ , which, in the

continuum limit, becomes

$$1 = \frac{a^2}{I} \int \frac{d\vec{q}}{(2\pi)^2} \frac{1}{e^{\omega_q/k_B T_c} - 1}. \quad (6)$$

For non-interacting electrons in 2D,  $\chi_s(q) - \chi_s(0) = 0$  for  $q < 2k_F$  [22], where  $k_F$  is the Fermi wave vector. The spin wave analysis therefore predicts  $T_c = 0$ , in agreement with a recent conjecture extending the Mermin-Wagner theorem for RKKY interactions in a non-interacting 2D system [25].

The study of thermodynamic quantities in *interacting* electron liquids especially in 2D has attracted quite some interest recently with the goal to find deviations from the standard Landau-Fermi liquid behavior, such as non-analytic dependences on the wave vector [15, 16, 17, 18]. In particular, it was found [16] that the static non-uniform spin susceptibility  $\chi_s(q)$  depends *linearly* on the wave vector  $q = |\vec{q}|$  for  $q \rightarrow 0$  in 2D (while it is  $q^2$  in 3D). This non-analyticity arises from the long-range correlation between quasiparticles mediated by virtual particle-hole pairs. Since the integral in Eq. (6) is dominated by the low  $q$ -behavior, one may replace  $\omega_q$  by its low- $q$  limit which turns out to be linear in  $q$  (see below) [23]. The integral in Eq. (6) can then be performed easily, allowing us to express  $T_c$  in terms of the derivative of the spin susceptibility,

$$T_c = \frac{A^2 I}{2k_B} \sqrt{\frac{3I}{\pi n_s}} \left. \frac{\partial \chi_s(q)}{\partial q} \right|_{q \rightarrow 0}. \quad (7)$$

For non-interacting electrons,  $\delta\chi_s(q) = 0$  at low  $q$  and we recover  $T_c = 0$ , in accordance with the MWT.

Let us include now e-e interactions. To calculate  $\chi_s(q)$ , we start from the Bethe-Salpeter (BS) equation for the two-body scattering amplitude [22]. Solving the BS equation formally, we can derive an exact and closed expression for the spin susceptibility given by

$$\chi_s(\vec{q}) = \frac{1}{L^2 D} \sum_{\vec{p}, \vec{p}'} \left( R(\vec{q}) \frac{1}{1 - \Gamma_{ir}^-(\vec{q}) R(\vec{q})} \right)_{\vec{p}\vec{p}'}, \quad (8)$$

where  $L = \sqrt{Na^2}$  is the system length,  $(\Gamma_{ir}^-)_{\vec{p}\vec{p}'}(\vec{q})$  the exact irreducible electron-hole scattering amplitude in the spin channel (see [22]),  $R_{\vec{p}}(\vec{q}) = -2iG(\vec{p} + \vec{q}/2)G(\vec{p} - \vec{q}/2)$  is the electron-hole bubble where  $G(\vec{p})$  is the exact propagator and  $\vec{p} \equiv (p_0, \vec{p})$  is the (D+1)-momentum with  $p_0$  the frequency. We have used a matrix notation in Eq. (8) where the indices run over  $\vec{p}$  ( $R$  is a diagonal matrix). Unfortunately,  $\Gamma_{ir}^-$  cannot be calculated exactly and some drastic approximations are required. The approximation we use consists in replacing the exact irreducible electron-hole scattering amplitude  $(\Gamma_{ir}^-)_{\vec{p}, \vec{p}'}$  by an averaged value calculated with respect to all possible values of  $p$  and  $p'$  near the Fermi surface, therefore we assume  $(\Gamma_{ir}^-)_{\vec{p}, \vec{p}'} = \Gamma_{ir}^-(\vec{q}) \forall p, p'$  [26].

Let us now put  $q_0 = 0$  (and suppress the  $q_0$ -argument from here on) and consider a  $q$ -independent short-ranged

(screened) interaction potential, yielding  $\Gamma_{ir}^-(\vec{q}) = -U$ . This allows us to derive from Eq. (8) a simple formula for  $\partial\chi_s/\partial q$  given by

$$\frac{\partial \chi_s}{\partial q}(q) = \frac{\partial \Pi(q)}{\partial q} \frac{1}{(1 + U\Pi(q))^2}, \quad (9)$$

where  $\Pi(q) = \sum_{\vec{p}} R_{\vec{p}}(q)/L^D$ . In the  $q \rightarrow 0$  limit, one can approximate the term  $\Pi(q)$  in the denominator of Eq. (9) by  $\chi_0(0) = -N_e$ . The resulting factor  $1/(1 - UN_e)^2$  in Eq. (9) can be interpreted as a type of random phase approximation (RPA) for the electron-hole scattering amplitude [27]. The corrections to the polarization bubble  $\Pi(q)$  (dominated by the first bubble correction to the self-energy) have been calculated in second order in perturbation theory (in  $U$ ) at small  $q$  by Chubukov and Maslov [16]. The result of this perturbative approach is  $\delta\Pi(q) = \Pi(q) - \Pi(0) \approx -4q\chi_s(0)\Gamma_s^2/3\pi k_F$ , where  $\Gamma_s \sim -Um^*/4\pi$  denotes the backscattering amplitude. When  $UN_e \ll 1$ , we recover from Eq. (9) the known result  $\delta\chi_s(q) = \delta\Pi(q)$  [16].

Now we are ready to obtain an estimate for the Curie temperature  $T_c$ . Replacing  $\chi_s(0)$  in  $\delta\chi_s(q)$  by its non-interacting limit  $\chi_0(0)$ , and assuming  $\Gamma_s = O(1)$  (this is an upper bound because  $\Gamma_s$  is a small parameter controlling the perturbation theory), we obtain then from Eq. (7)  $T_c \sim 25 \mu K$  for typical 2DEG parameters. This value of  $T_c$  becomes further enhanced by a numerical factor (e.g. of order 5 for  $r_s \sim 8$  [22]) if one uses an effective renormalized value for the spin susceptibility  $\chi_s = \chi_s(0)$  instead of  $\chi_0(0)$ . Though  $T_c$  is still rather small, it is now finite, confirming our arguments related to the Mermin-Wagner theorem that e-e interactions increase the Curie temperature. When  $UN_e$  is no longer negligible compared to 1,  $T_c$  is even further enhanced by an additional numerical factor  $1/(1 - UN_e)^2$  (see Eq. (9)). Close to the ferromagnetic Stoner instability of the electron system, reached when  $UN_e \sim 1$ , the Curie temperature  $T_c$  for the nuclear system is dramatically enhanced as could have been anticipated.

In the preceeding paragraphs, we replaced  $\Gamma_{ir}^-(q)$  by a  $q$ -independent constant operator. One can use instead another approximation called the local field factor approximation (LFFA). The idea of the LFFA is to replace the average electrostatic potential by a local field potential seen by an electron with spin  $\sigma$  (see [22] for a review). In this scheme  $(\Gamma_{ir}^-(q))_{pp'} \approx -V(q)G_-(q)$ , where  $G_-(q)$  is a local field factor and  $V(q) = 2\pi e^2/\kappa q$  the bare *unscreened* Coulomb interaction ( $\kappa$  is the dielectric constant). Within this approximation scheme the static spin susceptibility  $\chi_s$  becomes

$$\chi_s(q) = \frac{\chi_0(q)}{1 + V(q)G_-(q)\chi_0(q)}. \quad (10)$$

Determining precisely  $G_-(q)$  for all  $q$  is still an open issue. However, the asymptotic regimes are quite well established nowadays [22]. A semi-phenomenological interpolation formula based on the original Hubbard local

field factor [28] and modified in such a way that the compressibility sum rule is exactly satisfied reads [22, 29]:

$$G_-(q) \approx g_0 \frac{q}{q + g_0(1 - \chi_P/\chi_S)^{-1}\kappa_2}, \quad (11)$$

where  $g_0$  is related to the probability of finding two electrons (of opposite spins) at the same position in the electron liquid,  $(g\mu_B)^{-2}\chi_P$  is the Pauli susceptibility and  $\mu_B$  the Bohr magneton. For non-interacting electrons  $\chi_P/\chi_S = 1$ . An approximate form for  $g_0$  giving good agreement with quantum Monte Carlo (QMC) calculations has been proposed recently by Gori-Giorgi *et al.* [30]:  $g_0(r_s) \approx (1 + Ar_s + Br_s^2 + Cr_s^3)e^{-Dr_s}/2$ . In a 2DEG,  $r_s = 1/\sqrt{\pi n_e}a_B^*$  where  $a_B^* = \kappa/m^*e^2$  is the effective Bohr radius. The parameters  $A = 0.088$ ,  $B = 0.258$ ,  $C = 0.00037$ ,  $D = 1.46$  are fitting parameters reproducing QMC results for the 2DEG [30]. From Eqs. (7) and (10), one can easily determine  $T_c$  within the LLFA scheme to be given by

$$T_c = \frac{IA}{2k_B} \sqrt{\frac{3I}{\pi}} \frac{A}{(\alpha - 1)^2 g_0 V(a)}, \quad (12)$$

where  $\alpha = (1 - \chi_P/\chi_S)^{-1}$  and  $V(a)$  is the Coulomb potential evaluated at the interatomic distance  $a$ . The energy scale  $(\alpha - 1)^2 g_0 V(a)$  can be interpreted as a renormalized screened potential due to collective interaction effects that are incorporated in the LLFA. The ratio  $A/(\alpha - 1)^2 g_0 V(a)$  can be regarded as the small parameter of our theory. Quite remarkably, the LLFA predicts an exponential enhancement of  $T_c$  with increasing interaction parameter  $r_s$ . For a value of  $r_s \sim 5$ , this

theory already predicts a large  $T_c \sim 25$  mK, a temperature which is routinely achieved nowadays. Obviously, for some value of  $r_s$ , the dimensionless parameter  $A/(\alpha - 1)^2 g_0 V(a)$  exceeds unity. The truncation of the Schrieffer-Wolff transformation at lowest order becomes unjustified and feedback effects between the electron gas and the nuclear spins, not incorporated in our theory, become important. Nevertheless for relatively large values of  $r_s \lesssim 6$ , the condition  $A \ll (\alpha - 1)^2 g_0 V(a)$  is satisfied.

Although the spin wave analysis may overestimate  $T_c$ , the trend in all the approximation schemes we used is that e-e interactions increase dramatically the Curie temperature, possibly into the mK range for large  $r_s$  (therefore three orders of magnitude larger than  $E_{dip}$  which justifies our starting Hamiltonian). We note that the non-perturbative LLFA theory predicts higher  $T_c$ 's than the perturbative calculation in the short-ranged interaction. Finally, below  $T_c$ , the nuclear spins within the 2DEG polarize and generate an effective magnetic field of order of a few Tesla. This will create a small Zeeman splitting [31] in the 2DEG which should be detectable with e.g. optical or transport methods.

In summary, we have analyzed the Curie temperature  $T_c$  of nuclear spins in an interacting 2DEG using a mean field and a spin wave analysis. We have shown that electron-electron interactions considerably enhance the temperature for a ferromagnetic phase transition in the nuclear system, with  $T_c$  in the milli-Kelvin range for 2DEGs with  $r_s \sim 5 - 10$ . We thank B. Coish, L. Glazman, L. Kouwenhoven, and A. Yacoby for useful discussions. This work is supported by the Swiss NSF, NCCR Nanoscience, ONR, and JST ICORP.

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